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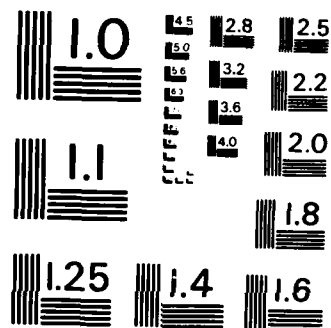
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Technical Report:

Stretching of Macromolecules in Turbulent Flows

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SUMMARY OF PREVIOUS AND ONGOING RESEARCH

In the following, we shall discuss the results obtained in the period September 1, 1984 to August 1, 1985 and complete the picture by a brief description of the ongoing work.

We have investigated the behaviour of dilute polymer solutions in flows which are strong in the sense that they can induce a significant deformation of the macromolecules. The rationale for this approach is that since turbulent flows are laminar on sufficiently small scales (such as the size of a polymer), the stretching of polymers by elongational flow bursts within the turbulent fluid can be studied by examining the behaviour of polymers in simple (laminar) elongational flows. The stretching, in turn, is a necessary condition for the flow to be significantly affected by the addition of minute quantities of polymer;¹ this is true regardless of the details of the mechanism leading to drag reduction (DR) and is a consequence of the observation that flow modification appears when the volume fraction of the polymers exceeds unity, i.e., when the polymers are strongly overlapping² (in a dilute solution this condition is satisfied only for stretched polymers).

We have investigated the stretching of polymers (the so called coil stretching transition³) in elongational flows using a combination of "physical" (scaling type⁴) and "chemical" (molecular level description in terms of local conformational states⁵) techniques. The results have been reported by us in References 6-14. Here, we summarize the ones that are the most important in the context of DR, and flow modification in general.

1. The flow parameter responsible for the stretching of polymers in elongational flows is the strain rate in the direction of the polymer. The critical strain rate (κ_c) at the coil stretching transition scales with the molecular weight of the polymer (M). The scaling law for random coiling polymers (PEO, PS, etc.) is

$$\kappa_c \sim \begin{cases} M^{-1.5} & \text{in } \Theta\text{-solvents} \\ M^{-1.6} & \text{in good solvents.} \end{cases} \quad (1)$$

Expanded polyelectrolytes (PAAM, NaPSS, etc.—at low salt concentrations) obey a different scaling law

$$\kappa_c \sim M^{-2}, \quad (2)$$

in all solvents. The above predictions are in excellent agreement with the experiments of the Bristol¹⁵ and Caltech¹⁶ groups. Inspection of expressions (1) and (2) indicates that the stretching of polyelectrolytes takes place at lower strain rates than that for the random coiling polymers. Thus, one expects that in flows of limited extensional strength, polyelectrolytes will be stretched and modify the flow more efficiently than random coiling molecules, but that the difference will disappear as the available strain rate levels are increased. The resulting prediction that the polyelectrolytes are better DR agents at low Reynolds numbers (R_e) but that the random coiling polymers become comparable or even better DR agents at the higher R_e , agrees with recent experimental observations.¹⁷

2. The stretching of polymers in elongational flows is subject to hysteresis, i.e., while relatively high strain rates are needed to stretch the polymer from its initial coil state, much smaller rates are needed to maintain it in the stretched state. This means that the events of stretching and flow modification need not be simultaneous in space and time; polymers which have been stretched while passing through a localized high strain rate region of the flow, will tend to maintain the stretched configuration and modify the flow at a considerable distance away from this region. While it is plausible that this nonlocal phenomenon can increase DR by providing more opportunity for flow modification, there is no experimental data which could clarify this issue (e.g., regarding the importance of hysteresis for DR).
3. The free-energy storage characteristics favor compact coil type polymers over the expanded ones since, in addition to the entropy loss, more energy can be stored in the conformational states of the coil-like polymers. Notice, however, that even if the energy storage mechanism was to play an important role in DR,¹⁸ (this issue is subject to controversy), our discussion of the random coiling polymers versus expanded polyelectrolytes indicates that the latter may be more efficient DR agents at the lower Reynolds numbers, and that the effect of the improved energy storage in the compact coils will become important only at higher R_e , at which the strain rates become high enough to stretch the coils.

Presently, our research proceeds simultaneously along two different directions:

1. We are investigating the effect of the coil stretching transition on the elongational flow field produced in a four-roll mill type of apparatus.¹⁶ Our calculations are based on the notion that in the region of space in which the polymers are completely stretched, they produce the dominant contribution to the stress field. The high elongational viscosity in this region tends to reduce the fluid strain rates to a level which is barely sufficient to maintain the polymers in their stretched state,¹⁹ thus producing the observed^{15,16} dip in the velocity profile (around the birefringent region in which the polymers are stretched).
2. We have obtained data from the NCAR group, which has computed the detailed two-dimensional turbulent velocity field based on a direct numerical solution (on a Cray 1 computer) of the Navier-Stokes equations.²⁰ We intend to analyze this data in order to study the (intermittent?) spatial and temporal distribution of the elongational flow regions, the R_e dependence of the elongational flow rates and the mechanisms leading to the formation of these regions (vortex-vortex interactions?²¹). Such an analysis may provide an essential link between our laminar flow results and turbulent flow phenomena and allow us to apply the former to the DR problem.



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DIRECTIONS OF FUTURE RESEARCH

The second phase of our research effort will start on March 1, 1986. The proposed direction of research in this period is based upon the results of our work in the first phase. In addition, it takes into account new results that were presented at the "Polymer-Flow Interaction" workshop which we organized in La Jolla on July 10-12, 1985. The most relevant results are summarized in the following:

1. Leal¹⁶ (Caltech)—Flow modification by stretched polymers can be observed in very dilute solutions. This disagrees with the previous results by the Bristol group¹⁵ which observed flow modification only at higher concentrations (essentially, of the order of the overlap concentration) and, presumably, is a consequence of the use of a more sensitive experimental technique (dynamic light scattering¹⁶ versus laser doppler velocimetry¹⁵).
2. Hoyt¹⁷ (SDSU)—DR had been observed at polymer concentrations as low as 0.02 ppm. The minute quantity of needed additive is the distinguishing feature of DR by polymers; other DR agents such as fibers, soaps or even sand, require much higher additive concentrations.
3. Hinch¹⁹ (Cambridge)—As the polymers get stretched by the local strain rate, their contribution to the fluid stress increases dramatically, resulting in a negative feedback that tends to reduce the strain rate in the fluid to the level where it is barely sufficient to maintain the polymers in their (almost) fully stretched state.

Another important consequence of the discussions at the "Polymer-Flow Interaction" workshop is the realization that progress in this area is inhibited by the lack of understanding of the fluid dynamics of orientable (rigid, "fiber"-like) and deformable ("polymer"-like) objects in flows that are strong enough to significantly alter the quiescent length and orientation distributions of those objects. That is not to say that the polymer part of the problem is well understood; on the contrary, while our understanding of polymer dynamics in quiescent solutions has grown dramatically in recent years due to the use of renormalization group^{22,23} and self-consistent field type²⁴ techniques, very little is known about polymer dynamics in strong, transient flows. For instance, while the "internal viscosity" of the macromolecule seems to play an important role in determining its response to such perturbations, its origin (and hence, its

dependence on the polymer parameters) remains unknown.¹⁴ However, even if one is willing to tremendously oversimplify the polymer description and model it as a non-linear dumbbell with a length-dependent friction coefficient,¹⁶ one is still unable to solve the non-Newtonian version of the Navier-Stokes equations when the strain rates in the fluid exceed the critical level at which the polymer molecules are strongly stretched. Thus, the phenomenon of flow modification by polymeric additives is much less well understood than its counterpart, e.g., the behaviour of polymers in a given strong flow. Since considerable experimental data is now available on both aspects of the polymer-flow interaction in laminar flows,^{15,16} and can be used to guide us in the construction of theoretical models, we believe that future research should address flow modification by the polymers as well as polymer distortion by the flow. Consequently, we propose that our second-phase research will proceed simultaneously along two different (but ultimately related) directions:

- A. Flow modification by polymeric additives, including (A.1) modification of laminar flows and (A.2) modification of turbulence and DR.
- B. Polymer deformation by flows including (B.1) internal viscosity, (B.2) polymer degradation and (B.3) polyelectrolyte effects.

In the following we present a more detailed statement of research along these lines.

A. Flow Modification by Polymeric Additives

A.1. Modification of Laminar Flows:

Modification of fluid flows by polymeric additives requires either large concentration or strong deformation of the polymers by the flow, the former case being traditionally associated with viscoelastic behaviour.²⁵ Since for our purposes (e.g., drag reduction¹⁷), we are more interested in the latter case, and since, as we have mentioned earlier, strong flow modification has been observed in laminar flows of dilute polymer solutions¹⁶, we shall restrict our attention to the dilute regime.

We shall continue the ongoing investigation of flow modification inside strong elongational flow devices (e.g., four-roll mills—see Figure 1). The theoretical investigation is based on the notion that the flow is appreciably modified only in the region where the polymers are almost fully stretched. Thus, as the polymers enter the

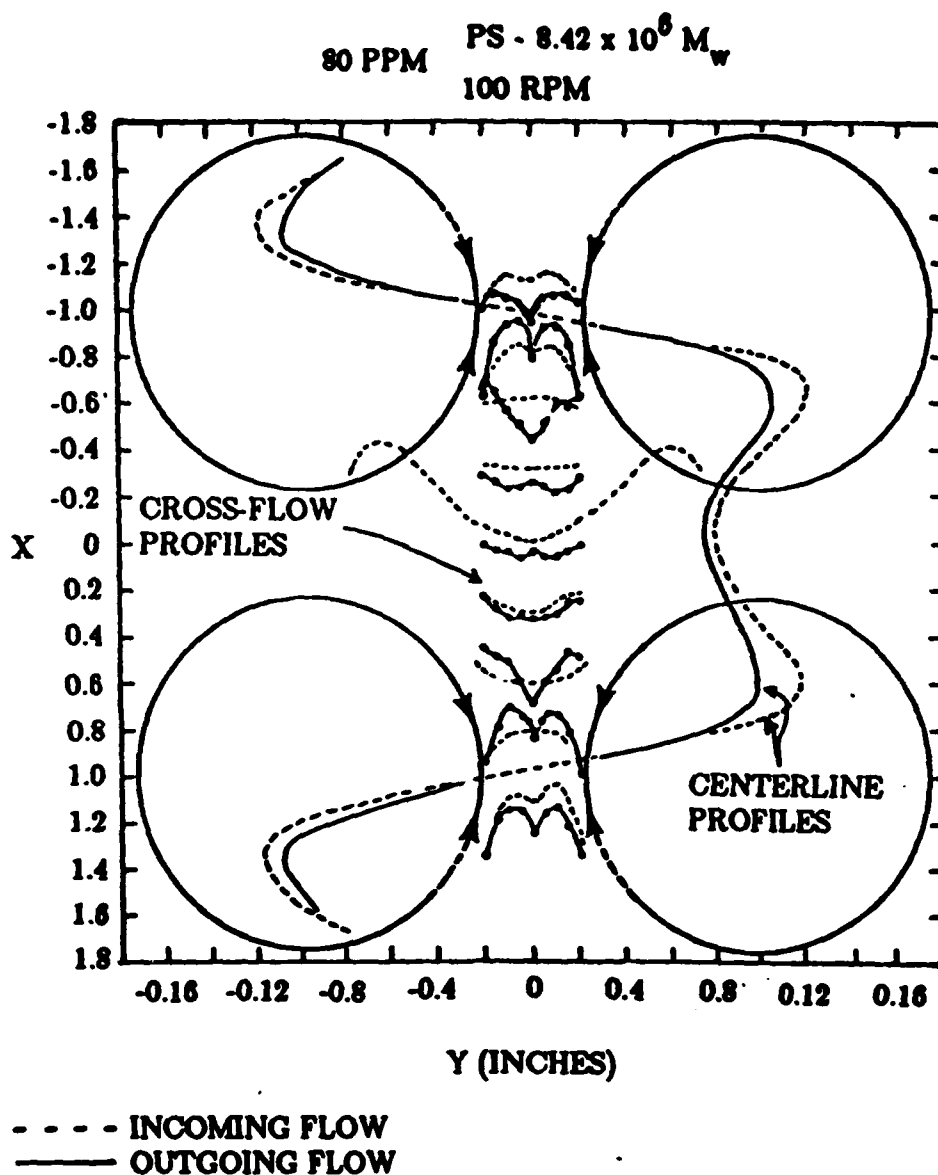


Fig. 4. Detailed velocity gradient profiles for an 80 PPM solution of polystyrene ($MW = 8.42 \times 10^6$) in Chlorowax; flow type $\lambda = 1$ and $\dot{\gamma} \sim 12 \text{ sec}^{-1}$. The curves labeled "centerline" show $\partial u / \partial z$ (or $\partial v / \partial y$) on the incoming (outgoing) axis, as a function of position as shown. The curves labeled "cross-flow" again show $\partial u / \partial z$ (or $\partial v / \partial y$) but now measured across the flow, at the position of the centerline value of the dashed (incoming) curve. The incoming and outgoing profiles are plotted along the same axis for comparison purposes. The incoming profiles are very similar to profiles with no polymer added.

elongational flow device, which operates at strain rates higher than the critical value, the coil configuration becomes unstable and the polymers are simultaneously advecting and deforming with the fluid element. They become fully stretched after a time

$$T = \frac{1}{2\kappa} \ln N \quad (3)$$

where κ is the strain rate and N the number of Gaussian segments (proportional to the molecular weight) of the polymer. Once this happens, the polymer contribution to the stress field increases dramatically and the elongational viscosity (η_{el}) becomes²⁶

$$\eta_{el} = \eta_s + \eta_p = \eta_s \left[1 + \frac{2\phi/3}{2 \ln \alpha + \ln(\pi/6\phi)} \right].$$

where η_s is the solvent (elongational) viscosity, α is the aspect ratio of a stretched polymer, and ϕ is the volume fraction (proportional to concentration) of the polymer. Notice that for typical values of polymer concentration (100 ppm) and molecular weight ($N = 10^4$), we get $\frac{\eta_p}{\eta_s} \approx 10^3 - 10^4$ and thus, in the region where the polymers are stretched, the elongational viscosity along the stretching axis is completely dominated by their contribution. We then separate the flow field into two regions (containing freely deforming and fully stretched polymers, respectively) and attempt to solve the resulting Navier-Stokes equations in both regions and then match the solutions at the boundary. This procedure will give us both the velocity profiles and the geometries of the two regions which will then be compared with experiment (see Figure 1).

At this stage, the interaction with experimentalists (e.g., Leal at Caltech and Keller in Bristol) will proceed both ways; for example, we anticipate that our theoretical model will give scaling-type (with powers of molecular weight of the polymer) predictions for the velocity profiles, which can then be tested experimentally.

The simple model presented above suffers from the restriction that strong flow modification is assumed to take place only in the region where the polymers are fully stretched. In reality, some flow modification will be present already for partially stretched polymers and, in addition, the resistance of the coiled polymers to stretching has to be taken into account in order to properly describe the behaviour at smaller strain rates. These modifications will be introduced into our model in a later stage in

an attempt to provide quantitative predictions for comparison with experiment. Another important extension of our research effort will be to investigate the case of strong, non-potential flows of non-zero vorticity, based on the hypothesis¹⁶ that only the strain rate in the direction of the polymer contributes to the stretching. Thus, we shall attempt to compute the flow modification in the case of a general steady two-dimensional flow and compare our results with experiments on flows of dilute polymer solutions in a two-roll mill type of apparatus;¹⁶ this is important since the above flow field has some of the characteristics of the vortex-vortex interaction processes that take place in turbulent flow and that are hypothesized to play a role in the stretching of polymers²¹ (and hence, in DR).

A.2. Turbulence Modification by Polymeric Additives

Just as in the laminar case, our proposed investigation of turbulence modification by polymers will be a continuation of an ongoing research effort²⁷ which focuses on one of the aspects of the polymer-flow interaction, namely, on the stretching of macromolecules in a given turbulent flow and neglects flow modification by the stretched polymers. To this end, we shall use the numerical data of McWilliams²⁸ who had mapped the two-dimensional velocity field in fully developed turbulence, and input it in the orientation and deformation equations for the polymers which, for simplicity, will be described as two-headed (uniaxial) vectors of variable length.²⁹ At this stage we will consider applying the same method to the recent results of three-dimensional simulations of turbulence performed at the Lawrence Livermore Laboratory,²⁷ in order to study the interaction between polymers and pure 3-D phenomena such as vortex stretching. This may be important in view of the correlations which were shown to exist between DR and the so-called "bathtub vortex inhibition" phenomenon,³⁰ which indicate that inhibition of vortex stretching by polymeric additives may be ultimately related to the microscopic mechanism of DR.

In order to determine the effects of the polymers on the fully developed turbulent flow, we will use renormalization group techniques. These techniques have been applied to ordinary turbulence by a number of workers. Important recent work is by Kraichnan,³¹ Siggia,³² and Orszag,³³ among others. We will extend the work of these authors to the case of turbulence modified by polymers. In particular, an anisotropic space-dependent stress tensor must be included—anisotropic to allow the anisotropy of

the polymer orientations to act, and space-dependent to allow for the polymers to be coils in some regions and stretched in other regions.

The idea of the renormalization group involves a way of understanding the relationship between very different scales in a complicated flow. In our case we want to know how the large scale flow in a pipe or around a ship is modified, assuming we know how the very small, laminar scale is modified by the polymer additive. In using the renormalization group, one assumes that this influence is not direct, but proceeds through the intermediate scales. The hard part of the calculation is to find how the parameters on one scale affect those at the next larger scale, and the easy part is to iterate from small scales up to large scales.

The primary aim is to find out the dependence of energy dissipation on the polymer concentration and molecular weight. We will compare the drag, expressed as power loss, at several different concentrations of polymers starting at zero, but staying within the dilute regime. Our results can then be compared with turbulent drag reduction experiments.

We expect that these calculations will be largely numerical, although some attention will be paid to the possibility of analytic results. We are more concerned with making our renormalization group equations as realistic as possible than with making them analytically tractable. Calculations will be done on our MicroVAX-II.

This approach is rather general. With little additional effort we can modify the polymer parameters. Thus, for example, we can treat the effect of rigid fibers rather than flexible polymers. In the long run, we may even be able to study other non-Newtonian flows including non-dilute polymer solutions.

B. Polymer Deformation in Strong Flows

In this part we shall focus on the role played by the polymer parameters in determining polymer behaviour in strong flows. In our view, the main problems related to the effect of polymer structure on the polymer-flow interaction are:

B.1. Internal Viscosity

This effect increases the resistance of the polymers to rapid deformations and thus tends to limit the stretching of polymers in flow fields of limited (strain rate) strength. The origin of this effect is unknown at present although several mechanisms such as

dynamic excluded volume type friction⁴ and non-Gaussian corrections to local chain structure³⁴ have been proposed. Our recent results^{12,14} lead us to believe that the internal viscosity phenomenon is a consequence of the correlations between the motions of the polymer chain and that of the surrounding solvent (backflow effects⁴); these correlations are enhanced in the presence of a deformational flow of limited strength⁷ and are expected to increase the resistance of the polymer to further deformation. The above effect will be studied using a variant of the mode coupling method, adapted to internal viscosity calculations.³⁴ However, unlike Adelman and Freed³⁴ who had investigated the somewhat unphysical, free-draining polymer chains, we will be taking the non-free-draining nature and the flow effects into account by introducing preaveraged (with respect to the ensemble appropriate for partially stretched polymers) hydrodynamic interactions and flow-induced long range correlations between the monomers along the chain. Apart from its fundamental importance, such a study will enable us to predict the relation between polymer behaviour in strong, transient flows, and parameters such as molecular weight, stiffness, and solvent viscosity.

B.2. Polymer Degradation

Degradation plays a three-fold role in the context of the DR phenomenon. Its "positive" aspect is that it provides us with essentially the only way of probing the degree of polymer extension in turbulent flows; experimental studies of the relationship between degradation and DR are now in progress.³⁵

The negative aspects are that degradation may limit the efficiency of DR in circulating pipe flows and that it may be the factor limiting DR in the high Reynolds number regime. The important question in this context is whether DR and polymer degradation can be separately manipulated by maximizing the former without causing a significant increase of the latter. The answer to this question is unknown at present.

Another important problem arises in connection with the fact that while the existing theories of chain rupture are based on thermal activation type notions,^{3,15} the experiments^{15,35} involve strong *transient* deformations of the macromolecules, i.e., inherently non-steady-state phenomena. It is almost certain that for high enough strain rates the process is not thermally activated but rather has to proceed through the disappearance of the barrier to disassociation (e.g., to chain rupture). This means that higher strain rates than what is usually assumed are actually needed to produce

degradation in transient flow, a prediction that is in agreement with the surprisingly low level of degradation (a few orders of magnitude lower than predicted!) observed in the Alaskan pipeline.³⁶

We shall develop a molecular level theory that will enable us to compute the possibility of chain rupture and the size distribution of the degradation products, which are expected to peak at $1/2, 1/4, \dots$, etc. of the original chain length¹⁵ depending on the applied flow strain rate, the total strain (i.e. the finite transit time through the stretching region of the flow field), and polymer parameters such as molecular weight, flexibility, and monomer bond strength.

This will be done for both the slow (thermally activated) and fast (transient) regimes of degradation and the results will be compared with experiment.^{15,35}

B.3. Polyelectrolytes

Expanded polyelectrolytes (PAAM, etc.) are among the best DR additives known at present (at least in the low to moderate Reynolds number regime¹⁷). Polyelectrolytes are unique in the sense that their expansion factor can be readily controlled by varying the salt (counterion) concentration in the solvent; in deionized water one obtains almost fully stretched polyelectrolytes while at high salt concentrations they collapse into the random coil configuration. Their behaviour as DR agents changes correspondingly³⁷ from "fiberlike" (no sharp onset of DR) to "coil-like" (with onset of DR). Another interesting property is that solubility of polyelectrolytes can be dramatically affected by addition of ionic groups along the chain. For example, polystyrene becomes water-soluble at high sulphonation levels ($\geq 75\%$) which, combined with the fact that narrow molecular weight fractions of high molecular weight polystyrene are commercially available, makes it an attractive candidate for use in studies of the molecular weight-dependence of DR.

The theory of polyelectrolytes is in a very primitive stage. While modern polymer solution theories can account for the transition between their asymptotic (fully stretched and collapsed coil) states, the behaviour at intermediate salt concentrations is not well understood even in quiescent solutions.³⁸ Classical problems that remain unresolved are the residual concentration dependence of the intrinsic viscosity even at the lowest experimentally accessible polyelectrolyte concentrations³⁹ and the apparently free-draining behaviour of partially extended polyelectrolytes at low salt

concentrations, which can be deduced from the molecular weight dependence of the critical strain rate of the coil-stretching transition in elongational flow. Experiments have observed a jump from the non-free-draining scaling $M^{-3/2}$ at high salt concentrations, to the free-draining scaling of M^{-2} at low salt concentrations.⁴⁰ This jump cannot be the result of the effective stiffening of the chain since, due to the cancellation between the effects of having fewer statistically independent segments and that of the segments being larger, the stiffening will not affect the critical strain rate at the coil stretching transition.⁹

Our preliminary investigations⁴¹ have led us to believe that the present difficulties with the theory of polyelectrolytes are the consequence of the failure of the Debye-Huckel approximation which assumes a homogenous distribution of counterions in the solution.⁴² In the polyelectrolyte case, the distribution of ions in the solvent contained "inside" the polymer chain, is different from that "outside;" moreover, the size of the "inside" region (i.e. the expansion factor of the polyelectrolyte chain), is itself a function of the ion concentration. These phenomena may give rise to a drastic reduction of the screening of the electrolyte interactions; the presence of long range coulombic forces can then explain the anomalous concentration dependence of the intrinsic viscosity.

We shall investigate the above question using a combination of variational methods that were recently developed to treat charged particle liquids,⁴³ with a Flory mean-field type description of the elastic behaviour of the polymer chains.³⁹ Later, we shall use the mathematical analogy between the Coulomb problem and the Oseen-type hydrodynamic interaction⁴ (taking into account the tensorial nature of the latter), and apply the methods of Reference 43, in order to construct a theory which will be able to describe the counterion concentration dependence of the hydrodynamic interaction.

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